

## Mid-infrared fibre-based optical comb

**P Maddaloni, P Malara, G Gagliardi and P De Natale**

Consiglio Nazionale delle Ricerche—Istituto Nazionale di Ottica Applicata,  
and European Laboratory for Nonlinear Spectroscopy (LENS) Comprensorio  
'A.Olivetti', Via Campi Flegrei 34, 80078 Pozzuoli (Naples), Italy

E-mail: [maddaloni@ino.it](mailto:maddaloni@ino.it)

*New Journal of Physics* **8** (2006) 262

Received 2 August 2006

Published 2 November 2006

Online at <http://www.njp.org/>

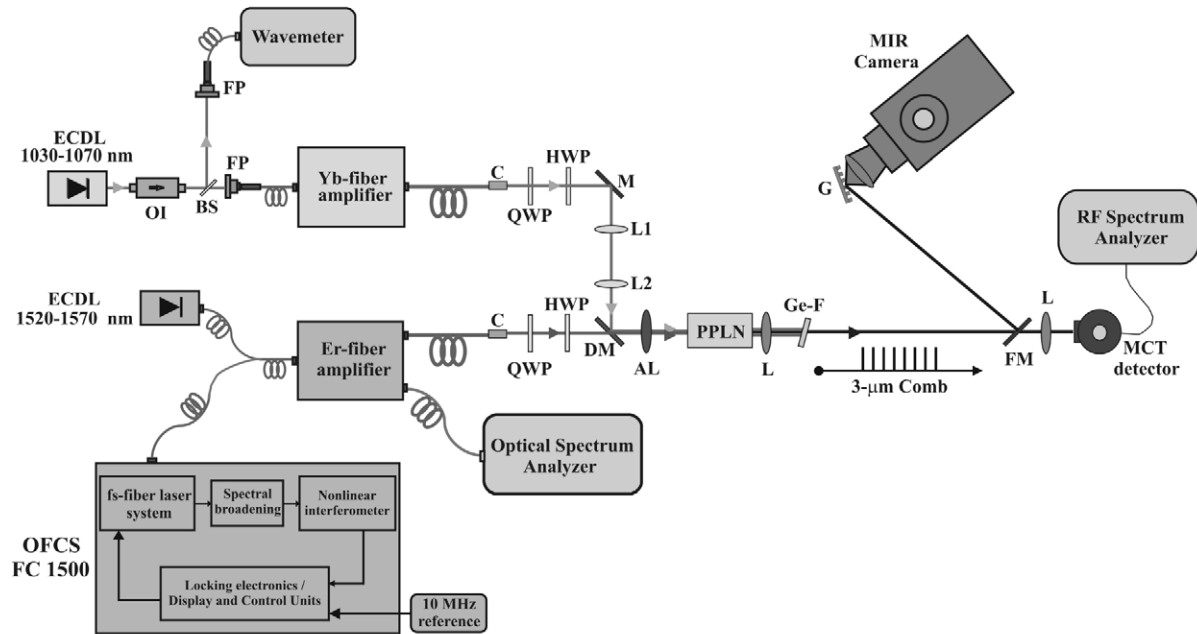
doi:10.1088/1367-2630/8/11/262

**Abstract.** An optical frequency synthesizer (OFS), based on difference-frequency generation (DFG), is demonstrated at  $3\ \mu\text{m}$ . By mixing a near-infrared (IR) OFS and a tunable continuous-wave (CW) laser in a periodically poled lithium-niobate (PPLN) crystal, a new comb is created from  $2.9$  to  $3.5\ \mu\text{m}$  (in  $180\ \text{nm}$  wide spans) with a  $100\ \text{MHz}$  mode spacing. By phase-locking, the pump laser to the near-IR OFS, the mid-IR comb is directly linked to the Cs primary standard via a global positioning system (GPS) time receiver. Then, the compactness of our fibre-based set-up opens real perspectives for the realization of transportable mid-IR frequency standards.

The introduction of mode-locked femtosecond (fs) lasers as a tool for synthesis and measurement of optical frequencies has suddenly led to new advances for metrological applications in the visible and near-IR spectrum [1]–[3]. Among these, the most prominent are in ultra-high resolution spectroscopy [4], inter-comparison of optical frequency standards [5], wavelength-multiplexed optical communication systems as well as multicolour interferometric length measurements [6]. In addition, their use is leading to new time-domain applications based on precise control of the carrier-envelope phase [7].

However, to fully benefit from the great potential of optical frequency synthesizers (OFSs), their spectral extension is crucial. Optical combs in the UV have already been demonstrated by high-order harmonic up-conversion, allowing for accurate metrology of atom transitions [8]–[10].

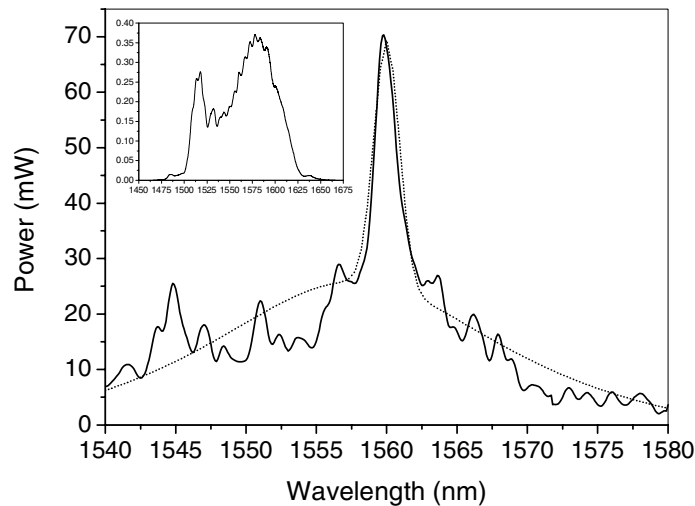
On the other hand, a direct comb synthesis in the IR can be a real breakthrough for absolute frequency metrology of molecular spectra. In this frame, a few schemes have been devised for the extension of OFSs to the IR region. Broadening the spectrum of fs mode-locked lasers through highly nonlinear optical fibres has already succeeded in extending combs up to  $2.3\ \mu\text{m}$



**Figure 1.** Schematic of the experimental apparatus: OI = optical isolator, BS = beam splitter, FP = fibre port, C = collimator, QWP = quarter wave plate, HWP = half wave plate, M = mirror, L = lens, DM = dichroic mirror, AL = achromatic lens, Ge-F = germanium filter, FM = flipping mirror, G = diffraction grating. The 3  $\mu\text{m}$  frequency comb is created by DFG in a PPLN crystal between a near-IR OFS and a cw laser. A fast, 100  $\mu\text{m}$  diameter MCT detector and a cryogenic InSb camera are used to characterize the generated mid-IR comb.

wavelength [11]. A 270 nm span frequency comb at 3.4  $\mu\text{m}$  has been realized by difference-frequency generation (DFG) between two spectral peaks emitted by a single uniquely designed Ti:Sa fs laser [12]. In a different approach, the metrological performance of OFSs has been transferred to the mid-IR, around 4.2  $\mu\text{m}$ , by phase-locking the DFG pumping sources to the teeth of a near-IR comb [13]. Such a method, however, precludes the possibility of direct comb-referencing for laser sources not based on frequency mixing, such as quantum cascade lasers.

In this paper, we demonstrate a frequency comb working around 3  $\mu\text{m}$ , based on a DFG process between a continuous wave (cw) laser source and a near-IR fibre-based OFS. The flexible character of our approach allows for a full and direct control of the comb features by simply acting on the parameters of the cw laser. In particular, by tuning its wavelength, the mid-IR comb covers the region from 2.9 to 3.5  $\mu\text{m}$  in 180 nm wide spans. A co-propagating, cw DFG beam with a power of several milliwatts is simultaneously produced in order to characterize the generated comb. The experimental apparatus for the creation of the 3  $\mu\text{m}$  frequency comb is shown in figure 1. The DFG signal radiation comes from a near-IR OFS (FC1500, Menlo Systems) covering the octave from 1050 to 2100 nm. Its repetition rate (100 MHz) and carrier-envelope offset frequency are locked to a reference oscillator based on a high-stability GPS-disciplined 10 MHz quartz, including a Rb-clock. The signal beam is then provided by feeding a fraction (25 mW) of the fs fibre laser system output (before the spectral broadening stage), covering



**Figure 2.** Power spectral distribution of the near-IR comb at the output of the EDFA, as recorded by the OSA with a resolution of 0.1 nm. The area under the curve gives the power of the DFG signal source (0.7 W). The dotted line is a fit of a parabola plus a Gaussian function to the experimental curve and is used in the theoretical model to predict the power of the generated  $3\ \mu\text{m}$  comb (see text). The inset also shows the power spectrum coming from the near OFS which initially seeds the EDFA.

the 1500–1625 nm interval, to an external Er-doped fibre amplifier (EDFA). The power spectral distribution, resulting from the convolution with the EDFA gain curve, is recorded by an optical spectrum analyser (OSA) and shown in figure 2 together with the best fitting curve (i.e. the sum of a parabola and a Gaussian function). The amplified comb beam has an average power of 0.7 W and spans from 1540 to 1580 nm (with a 100 MHz spacing), corresponding to nearly  $N_t = 50\ 000$  teeth in the frequency domain. Concerning the intensity and frequency noise spectral density, simultaneous amplification of many optical carriers does not introduce any appreciable additional noise in the process, owing to the slow gain dynamics which makes the amplifier practically immune from crosstalk effects [14]. Actually, use of advanced fibre-based devices, which benefit from the continuous progress in telecom technology, may give additional advantages for the realization of more and more effective set-ups.

The pump beam is generated by an external-cavity diode laser (ECDL) emitting in the range 1030–1070 nm and is amplified by an Yb-doped fiber amplifier which delivers up to 0.7 W, preserving the linewidth of the injecting source (less than 1 MHz). Then each beam passes through a quarter-wave plate, to correct for residual ellipticity, followed by a half-wave plate to get an s-polarized beam (parallel to the crystal optical axis), as required by the quasi phase matching (QPM) condition. The two laser beams are finally combined on to a dichroic mirror and focused by a near-IR achromatic lens ( $f = 100\ \text{mm}$ ) into a temperature-controlled, antireflection-coated, periodically poled lithium-niobate (PPLN) crystal. The latter consists of an array of nine channels, with different poling periods ranging from 29.6 to 30.6  $\mu\text{m}$ . Once the wavelength of the pump source is fixed (1055 nm), the channel and temperature value (between 50 and 80° C) are properly chosen to satisfy the QPM condition with the centre wavelength of the near-IR comb (1560 nm). The teeth on both sides are involved in as many DFG processes, with a signal power described by the envelope (dotted line) represented in figure 2, and with a

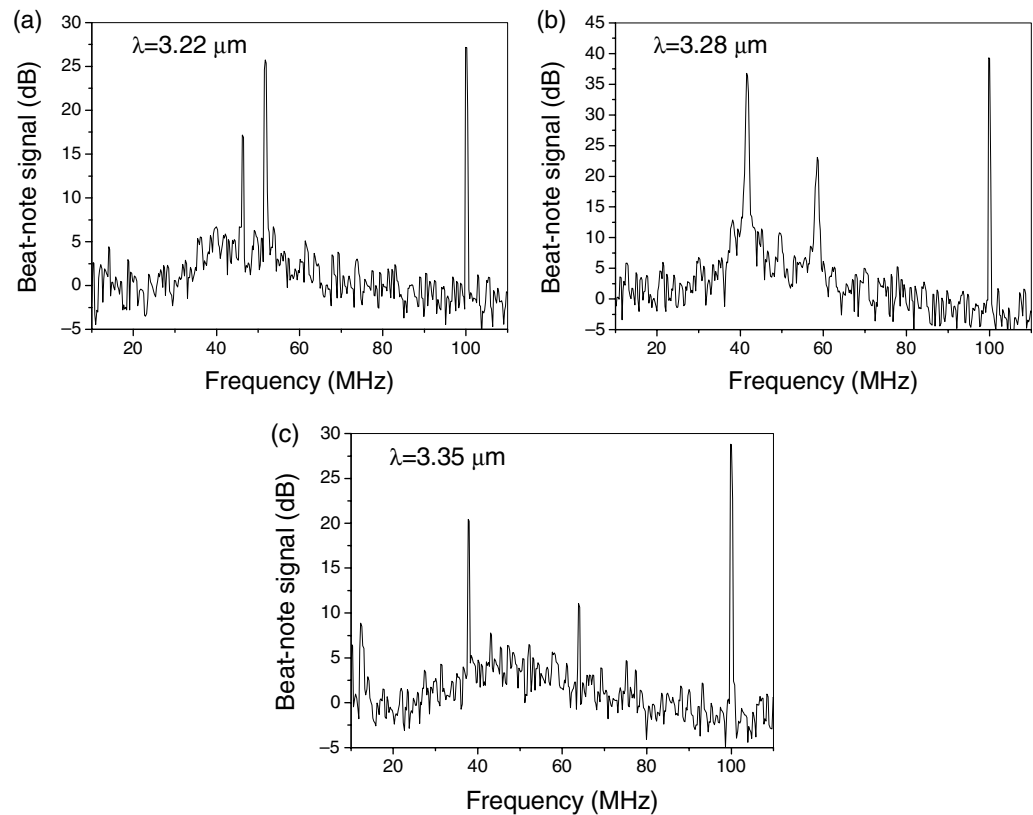
conversion efficiency decreasing according to the well-known  $\text{sinc}^2$  law [15]. Then, the two-wave DFG theory (in the plane-wave approximation) is easily generalized to the case of one pump frequency and many signal frequencies ( $N_t = 50\,000$ ), predicting a value of about  $20\ \mu\text{W}$  for the power integrated over the whole comb wavelength span.

The  $3\ \mu\text{m}$  comb is detected by filtering the DFG idler beam from the unconverted near-IR light and focusing it on to a liquid-nitrogen-cooled, 150 MHz bandwidth HgCdTe (MCT) detector. In this way, a radio-frequency (rf) beat note at  $\omega_r = 100\ \text{MHz}$  is recorded by a spectrum analyser, which is the sum of the beat signals between all pairs of consecutive teeth in the generated comb. The latter has a bandwidth of 180 nm (5 THz) centred near  $3.3\ \mu\text{m}$  and its measured overall power  $P$  is about  $5\ \mu\text{W}$ , the slight discrepancy with the above theoretical value being imputable to the Gaussian nature of the beams. This value corresponds to a power of nearly  $5\ \mu\text{W}/N_t = 100\ \text{pW}$  per mode of the IR comb. Since the linewidth of the ECDL is around 1 MHz, the DFG comb lines are significantly wider than those of the near-IR OFS. This can be overcome either by locking the ECDL to an external high-finesse Fabry-Perot cavity or to a tooth of the near-IR comb.

In most schemes, the metrological performance of an OFS is usually transferred to a cw laser by phase-locking the rf beat note between the laser and the closest tooth in the comb to a local oscillator by feeding appropriate frequency corrections back to the laser itself. Our experiment is also intended to demonstrate that such a scheme is possible in a hardly accessible spectral region. For this purpose, a cw DFG beam is simultaneously produced by the apparatus in figure 1.

This is accomplished by simultaneously seeding the Er-fibre amplifier with an ECDL emitting in the 1520–1570 nm interval (having a linewidth less than 500 kHz). In this configuration, a cw  $1.5\ \mu\text{m}$  beam is also produced by the EDFA and the emission spectrum of the EDFA is modified. This unavoidably reduces the power of the amplified comb beam and consequently that of the  $3\ \mu\text{m}$  DFG comb ( $P = 1\ \mu\text{W}$ ). It should be noted that this is a consequence of the further amplification process, only used for characterization. The cw signal beam gives rise to a second DFG process with the pump radiation thus producing a cw idler beam around  $3\ \mu\text{m}$  with a power between 1.5 and 3 mW, depending on the wavelength. As a consequence, two additional rf beat notes at  $\omega_1 = \omega_{\text{cw}} - \omega_n$  and  $\omega_2 = \omega_{n+1} - \omega_{\text{cw}}$  are detected between the DFG cw radiation at  $\omega_{\text{cw}}$  and its two closest IR comb teeth at  $\omega_n$  and  $\omega_{n+1}$  respectively (see figure 3). The amplitude difference between these two peaks is likely caused by the non-flat AC responsivity of the detector preamplifier. In figure 3, the signal-to-noise ratio (SNR) for such beat notes can be measured as the  $1.5\ \mu\text{m}$  ECDL wavelength is tuned from 1540 to 1570 nm ( $\omega_{\text{cw}}$  from  $3.22$  to  $3.35\ \mu\text{m}$ ). The SNR value reaches a maximum of 35 dB at the centre wavelength (1555 nm), while decreases almost symmetrically down to less than 20 dB at the upper and lower edges. This limits to about 130 nm the interval which is generally suitable for use in optical phase-locked systems. Actually, the 180 nm span can be fully exploited as higher beat notes are expected when an external  $3\ \mu\text{m}$  mW power source is used (i.e. in absence of the simultaneous cw DFG beam coming from the same EDFA). Moreover, SNR levels can be further improved selecting a smaller number of teeth by using a diffraction grating.

Finally, by tuning the  $1\ \mu\text{m}$  laser wavelength, the centre frequency of the DFG comb is tuned from  $3.1$  to  $3.4\ \mu\text{m}$ , without any need to adjust the QPM conditions. This is shown in figure 4 where the peak signal of the beat note at 100 MHz is plotted as a function of the  $1\ \mu\text{m}$  wavelength, the upper limit being set by the laser tunability range. By also tuning the QPM

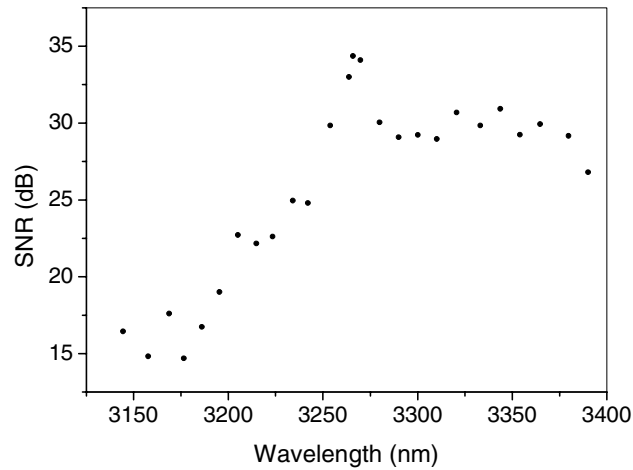


**Figure 3.** Beat signals recorded by the rf spectrum analyser at the centre (b) and at the ends ((a) and (c)) of the  $3 \mu\text{m}$  comb span. The peak at  $\omega_r = 100 \text{ MHz}$  is the sum of the beat signals between all pairs of consecutive teeth in the generated comb, while the peaks at  $\omega_1$  and  $\omega_2$  correspond to the beat notes between the DFG cw radiation and its two closest comb teeth. Asymmetry between these two peaks is mostly due to the non-uniform frequency response of the detector preamplifier. Resolution and video bandwidth are, respectively, 10 kHz for panels (a) and (c) and 3 kHz for panel (b). The sweep time for all the acquisitions is about 1 s.

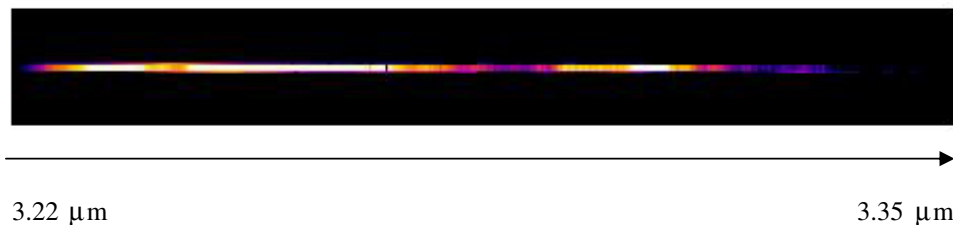
conditions, higher conversion efficiencies and further extension of the span (from  $2.9$  to  $3.5 \mu\text{m}$ ) can be accomplished.

The beam intensity profile of the generated mid-IR comb was also monitored by means of a liquid-nitrogen-cooled InSb camera consisting of an array of  $320 \times 256$  square pixels ( $30 \mu\text{m}$  size). For this purpose, the DFG beam was directed on to a reflective diffraction grating ( $133 \text{ lines mm}^{-1}$ ) and the first order (about 95% of the incident power) re-focused by a  $\text{CaF}_2$  lens onto the camera. The recorded beam pattern is shown in figure 5, where a very wide spectral span ( $3.22$ – $3.35 \mu\text{m}$ ) is shown. Pixel-to-wavelength calibration was obtained by imaging the cw DFG beam for two different wavelengths.

In conclusion, we have demonstrated an optical frequency comb directly operating around  $3 \mu\text{m}$ . DFG in a PPLN crystal between a near-IR OFS and a widely tunable cw laser has been used to transfer the frequency comb metrology to the  $2.9$ – $3.5 \mu\text{m}$  spectral window. A co-propagating cw DFG beam has been used to characterize the mid-IR comb, showing its potential for future



**Figure 4.** SNR for the beat note at  $\omega_r = 100$  MHz as a function of the DFG wavelength, recorded by tuning the pump source from 1040 to 1070 nm. Each point represents a frequency comb with a span of about 150 nm. The asymmetric behaviour with respect to the central wavelength is caused by the decrease in the optical power of the Yb fibre amplifier.



**Figure 5.** Mid-IR ‘rainbow’ recorded by the InSb camera (in false colours). The diffraction pattern is obtained by directing the  $3\ \mu\text{m}$  generated comb on to a grating with a first-order 95% reflectivity. Re-focusing by a lens allows us to observe in a single acquisition the whole comb span (3.22–3.35  $\mu\text{m}$ ).

metrological applications of novel lasers under development [16]. Of course, the demonstrated scheme can be similarly applied to different IR regions, provided that nonlinear crystals with suitable transparency and conversion efficiency are available. Furthermore, the generated mid-IR OFS can provide metrological performance traceable to the primary Cs frequency standard by phase-locking the pump laser to a tooth of the GPS-based near-IR comb [17]. Then, the richness of strong ro-vibrational transitions that characterizes the  $3\ \mu\text{m}$  region may open new interesting perspectives for high resolution molecular spectroscopy [18] as well as for high precision tests of fundamental physical theories [19]. In the time domain, the realization of phase-coherent mid-IR pulses offers a novel tool for Fourier-transform molecular spectroscopy [20] and coherent control of molecular reactions in a previously uncovered spectral region [21]. Moreover, by taking advantage of both the broad bandwidth and the outstanding frequency accuracy, the generated mid-IR comb may be used in interferometric set-ups to measure refractive indexes in a still quite unexplored spectral region, with very high precision and accuracy [22]. The

ability of imaging the generated mid-IR comb also envisages the extension of frequency-comb-based interferometric/olographic techniques, already demonstrated at  $1.5 \mu\text{m}$  [23], to the mid-IR region. Finally, the compactness and portability of both the near-IR OFS and our fibre-based DFG apparatus [24] paves the way to the realization of a really transportable mid-IR optical frequency comb.

## Acknowledgments

We acknowledge fruitful discussions with G Giusfredi and P Cancio. This research was partially funded by the MIUR within the FIRB project RBNE01KZ94 ‘Microdispositivi fotonici in Niobato di Litio’ and partially by the JRA contract RII3-CT-2003-506350 (FOSCIL).

## References

- [1] Reichert J, Holzwarth R, Udem T and Hänsch T W 1999 Measuring the frequency of light with mode-locked lasers *Opt. Commun.* **172** 59–68
- [2] Jones D J, Diddams S A, Ranka J K, Stentz A, Windeler R S, Hall J L and Cundiff S T 2000 Carrier envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis *Science* **288** 635–9
- [3] Udem T, Holzwarth R and Hänsch T W 2002 Optical frequency metrology *Nature* **416** 233–7
- [4] Holzwarth R, Udem T, Hänsch T W, Knight J C, Wadsworth W J and Russell P St J 2000 Optical frequency synthesizer for precision spectroscopy *Phys. Rev. Lett.* **85** 2264–7
- [5] Long-Sheng Ma, Bi Z, Bartels A, Robertsson L, Zucco M, Windeler R S, Wilpers G, Oates C, Hollberg L and Diddams S A 2004 Optical frequency synthesis and comparison with uncertainty at the  $10^{-19}$  level *Science* **303** 1843–5
- [6] Telle H R and Sterr U 2001 Generation and metrological application of optical frequency combs ‘*Frequency Measurement and Control*’ ed A N Luiten (Berlin: Springer) pp 295–313
- [7] Apolonski A, Poppe A, Tempea G, Spielmann C, Udem T, Holzwarth R, Hänsch T W and Krausz F 2000 Controlling the phase evolution of few-cycles light pulses *Phys. Rev. Lett.* **85** 740–3
- [8] Witte S, Zinkstok R T, Ubachs W, Hogervorst W and Eikema K S E 2005 Deep-ultraviolet quantum interference metrology with ultrashort laser pulses *Science* **307** 400–3
- [9] Jones R J, Moll K D, Thorpe M J and Ye J 2005 Phase-coherent frequency combs in the vacuum ultraviolet via high-harmonic generation inside a femtosecond enhancement cavity *Phys. Rev. Lett.* **94** 193201
- [10] Gohle C, Udem T, Rauschenberger J, Holzwarth R, Hermann M, Schüssler H A, Krausz F and Hänsch T W 2005 A frequency comb in the extreme ultraviolet *Nature* **436** 234–7
- [11] Thomann I, Bartels A, Corwin K L, Newbury N R, Hollberg L, Diddams S A, Nicholson J W and Yan M F 2003 420 MHz Cr : forsterite femtosecond ring laser and continuum generation in the  $1\text{--}2 \mu\text{m}$  range *Opt. Lett.* **28** 1368–70
- [12] Foreman S M, Marian A, Ye J, Petrukhin E A, Gubin M A, Muecke O D, Wong F N C, Ippen R P and Kaertner F X 2005 Demonstration of a HeNe/CH<sub>4</sub>-based optical melocular clock *Opt. Lett.* **30** 570–2
- [13] Mazzotti D, Cancio P, Giusfredi G, Prevedelli M and De Natale P 2005 Frequency-comb-based absolute frequency measurements in the mid-infrared with a difference-frequency spectrometer *Opt. Lett.* **30** 997–9
- [14] Cancio P, Zeppini P, De Natale P, Taccheo S and Laporta P 2000 Noise characteristics of a high-power ytterbium-doped fibre amplifier at 1083 nm *Appl. Phys. B* **70** 763–8
- [15] Maddaloni P, Gagliardi G, Malara P and De Natale P 2005 A 3.5 mW continuous-wave difference-frequency source around  $3 \mu\text{m}$  for sub-Doppler molecular spectroscopy *Appl. Phys. B* **80** 141–5
- [16] Capasso F, Gmachl C, Paiella R, Tredicucci A, Hutchinson A L, Sivco D L, Baillargeon J N, Cho A Y and Liu H C 2000 New frontiers in quantum cascade lasers and applications *IEEE J. S. T. Quantum Electron.* **6** 931–47

- [17] Fox R W, Diddams S A, Bartels A and Hollberg L 2005 Optical frequency measurements with the global positioning system: tests with an iodine-stabilized He–Ne laser *Appl. Opt.* **44** 113–20
- [18] Thorpe M J, Moll K D, Jones R J, Safdi B and Ye J 2006 Broadband cavity ringdown spectroscopy for sensitive and rapid molecular detection *Science* **311** 1595–9
- [19] Bellini M, De Natale P, Inguscio M, Varberg T D and Brown J M 1995 Precise experimental test of models for the breakdown of the Born–Oppenheimer separation: the rotational spectra of isotopic variants of lithium hydride *Phys. Rev. A* **52** 1954–60
- [20] Keilmann F, Gohle C and Holzwarth R 2004 Time-domain mid-infrared frequency-comb spectrometer *Opt. Lett.* **29** 1542–4
- [21] Cundiff S T and Ye J 2003 Colloquium: femtosecond optical frequency combs *Rev. Mod. Phys.* **75** 325–42
- [22] Zhang J, Lu Z H and Wang L J 2005 Precision measurement of the refractive index of air with frequency combs *Opt. Lett.* **30** 3314–6
- [23] Choi S, Yamamoto M, Moteki D, Shioda T, Tanaka Y and Kurokawa T 2006 Frequency-comb-based interferometer for profilometry and tomography *Opt. Lett.* **31** 1976–8
- [24] Maddaloni P, Malara P, Gagliardi G and De Natale P 2006 Two-tone frequency modulation spectroscopy for ambient-air trace gas detection using a portable difference-frequency source around 3  $\mu\text{m}$  *Appl. Phys. B* **85** 219–22